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MINERALS FROM GYÖNGYÖSOROSZI[1]

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In the most important coloured metal-ore mining district of Hungary, in Gyöngyösoroszi, in the country of Heves and to the north of this village, the ore lodes extend over an area limited by andesite inclusions in an exposed older pyroxene andesite agglomerate. The greater part of the lodes fill up longitudinal, the smaller part, transversed fissure-systems, most of the latter are already in the andesite area of the Mátra Mountains. The lodes are predominantly filled up by different kinds of quartz, the longitudinal lodes show owing to their rhythmical separation a partly cryptocrystalline, striated structure, whilst the transversed lodes are filled with coarsely crystalline quartz often exhibiting a brecciated structure. So far, it only seemed worthwhile to operate the lodes filled with striated quartz as merely they contain ore. The minerals described below were mainly found in the principal lode, the Károly lode and the Péter-Pál one.

Until quite recently only investigations were carried out in Gyöngyösoroszi, the exposure of the lodes actually only began when the adit was opened up. Now there is also more exposing than productive work, thus it is not surprising that the literature dealing with the minerals of the deposite is still very incomplete. From the geological and mining-geological point of view P. Rozlozsnik[1] and G. Pantó[2, 3], from the mineral one F. Papp[4], K. Sztrókay[5, 6, 7], as well as Koch—Mezősi—Grasselly[8] dealt with the subject.

The dominating ore mineral of the ore lodes is sphalerite, beside it galena can be found in considerable amounts. It is interesting, that among the ores of the Károly lode ZnS occurs not only as sphalerite, but also as wurtzite.

Wurtzite forms at lower temperature from solutions of acid character therefore, it occurs far less frequently among the primar ore lode minerals than sphalerite which forms from the only quite mildly acid or even alkaline solutions, over wide temperature ranges. If wurtzite can be found it can usually be observed among the most recently formed sulphide ores in the form of needle crystal aggregates and very rarely as individual overgrown small crystals. The latter are mostly sphalerite paramorphoses after wurtzite. In the oxidation zone owing to the action of acid solutions sometimes wurztzite crystal groups and sometimes single small crystals can be found. *In Gyöngyösoroszi würtzite belongs to the primarily formed sulphide ores.* Separate crystals could not be detected, they did not either

[1] Delivered as a lecture to the Geochemical Commission of the Hungarian Academy of Science at its Meeting in April 1954.

appear as »Schalenblende«, but as radial fanlike one cm long crystal groups attaining forming bands ingrown in quartz or band rows separated by thin quartz layers (Microphot. 1. and 2.). Its colour is clove brown, its lustre half metallic.

Considering that wurtzite and sphalerite are difficult to differenciate under the microscope thin sections of the ore were also prepared. In which the wurtzite crystal bundles were sometimes pointed and sometimes blunt, pyramidelike, transparent showing lighter or darker brownish-yellow colour. The colour changed from zone to zone depending on the amount of FeS. The cleavage according to (0001) could be observed on every sample examined. On certain points the crystal groups were quite free from inclusions; however, frequent by chalcopyrite inclusion rows extending along the boundaries of the crystal groups can be seen, on which dense chalcopyrite inclusion rows (Microphot. 3, 4 and 5) occur nearly perpendicularly, forming parallel with the cleavage plane usually rendering the wurtzite crystal group quite opaque. The substance of the chalcopyrite laths formed alternately with the wurtzite and intergrew with it in an oriented manner. In sections under the microscope with an oil immersion these orientally separated very abundant chalcopyrite inclusion rows running parallel with the cleavage plane can be well detected (Microphot. 6). Besides several of these micron sized inclusions wurtzite also contains oriented chalcopyrite sphenoid inclusions (Microphot. 7). As can be seen on microphoto No. 8. these sphenoids are the remnants of the chalcopyrite replaced by wurtzite.

Between crossed nicols the wurtzite crystals exhibit an anisotrope character showing an interference in the diagonal position. Optically they have a positive character. Thus it is beyond doubt that wurtzite and not a paramorph after wurtzite are involved. Even the light coloured crystals and crystals groups did not transform into sphalerite, although according to Ehrenberg the latter transform more rapidly than the dark coloured crystals containing much iron.

It is very interesting that on the (0001) plane of the wurtzite crystal groups sphalerite tetrahedrons overgrowing in an oriented manner can be found, i. e. tetrahedrons which sometimes do not contain any inclusions and others containing many chalcopyrite inclusions and even chalcopyrite sphenoids intergrown orientally with sphalerite (Microphot. 9 and 10). The most interesting sample is that on which on the (0001) plane of the wurtzite crystal group larger, incompletely developed sphalerite tetrahedrons are overgrown parallel whilst on the top of the sample a tetrahedron can be seen. The entire crystal group like most of our wurtzite crystal groups is embedded in quartz (Microphot. 11).

On the basis of its cleavage directions and owing to its isotropic character sphalerite can always be distinctly distinguished from wurtzite. Sometimes the sphalerite is lighter containing less dissolved FeS and sometimes darker than wurtzite. A remarkable section is that on which the fanlike wurtzite crystal group containing a large amount of FeS are overgrown as light sphalerite (Microphot. 12). In the interior of the sphalerite crystals associated with wurtzite the parts containing more and those containing less iron alternate in zones (Microphot. 13).

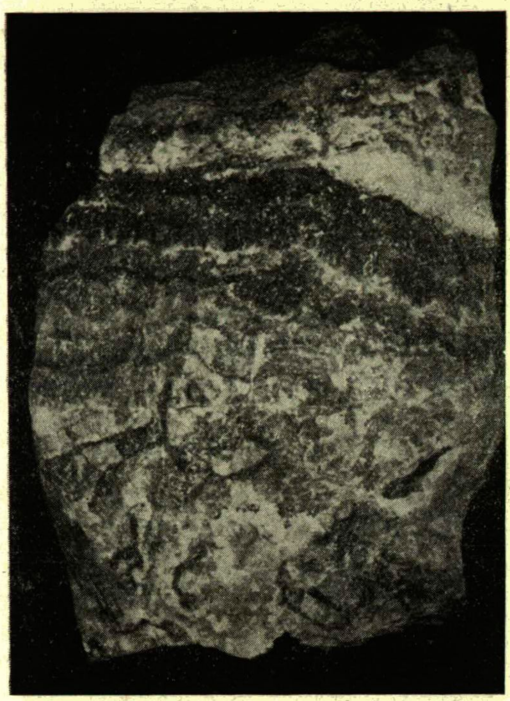

Fig. 1.
Lode fragment with wurtzite
about 1/3 the original size

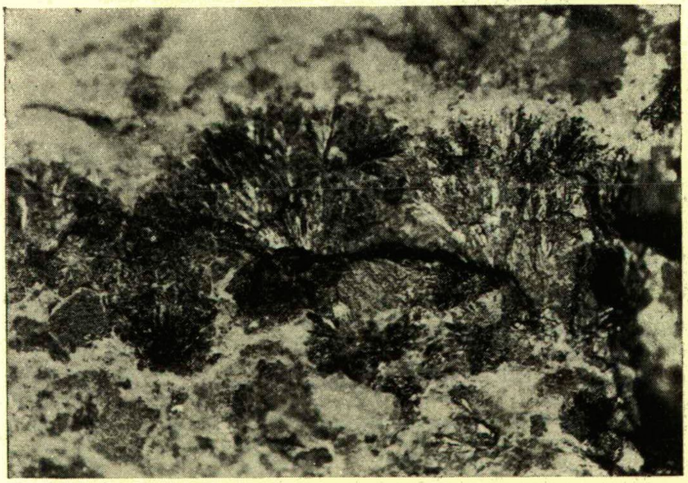

Fig. 2.
Lode fragment with wurtzite, $\times 4$

Fig. 3.
Zonal wurtzite with chalcopyrite inclusions, $\times$ 40

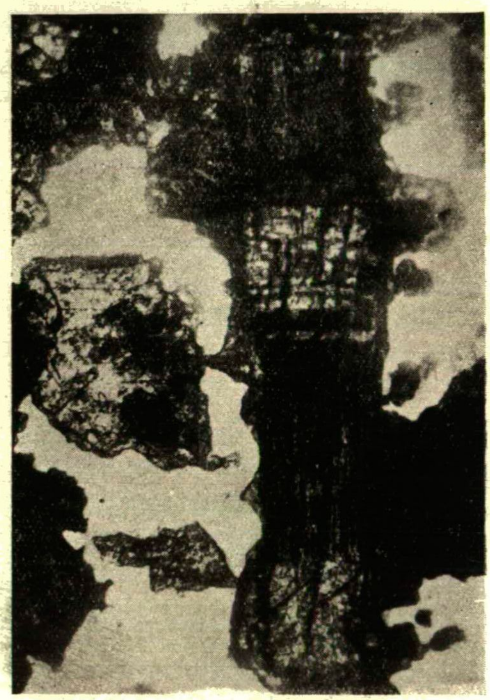

Fig. 4.
Wurtzite, with the 0001 plane parallel and to it perpendicular oriented chalcopyrite inclusion rows, $\times$ 90

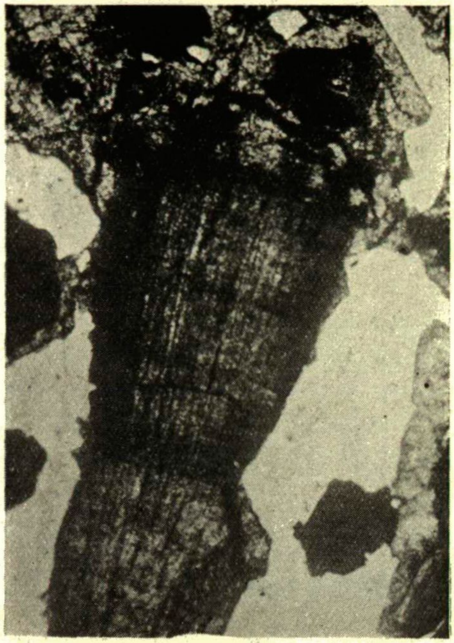

Fig. 5.
Zonal wurtzite, the section parallel to the *c* axis, $\times 110$

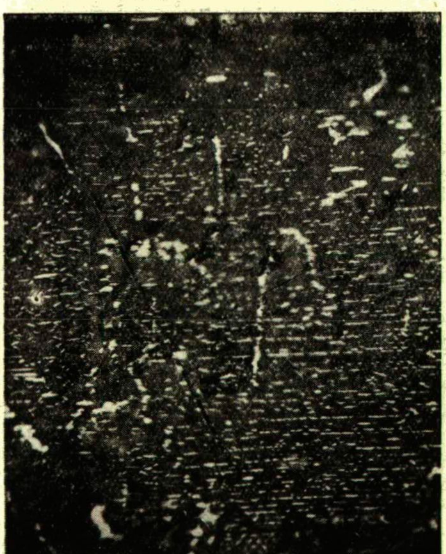

Fig. 6.
Wurtzite, chalcopyrite inclusion rows settled parallel with 0001, $\times 400$

Fig. 7.

Wurtzite with oriented chalcopyrite sphenoids, $\times 50$

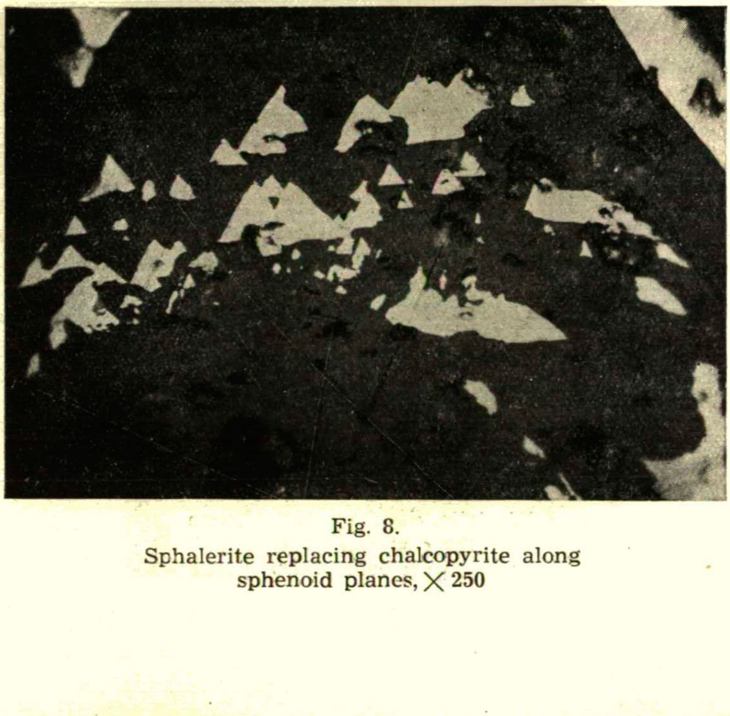

Fig. 8.

Sphalerite replacing chalcopyrite along sphenoid planes, $\times 250$

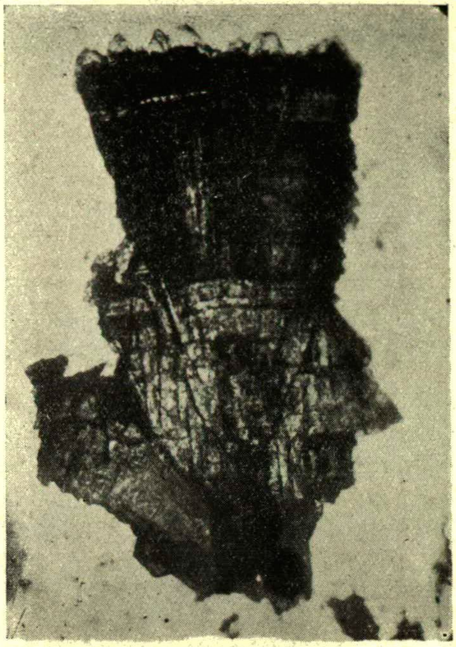

Fig. 9.
Zonal wurtzite crystal bundle, on the plane 0001 with sphalerite tetrahedrons, $\times$ 110

Fig. 10. Wurtzite, with sphalerite tetrahedron, $\times$ 150

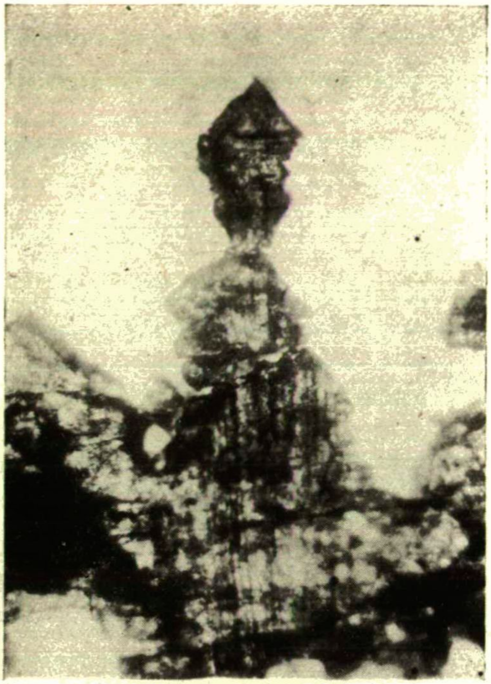

Fig. 11.
Wurtzite crystal bundle with oriented overgrown sphalerite, × 140

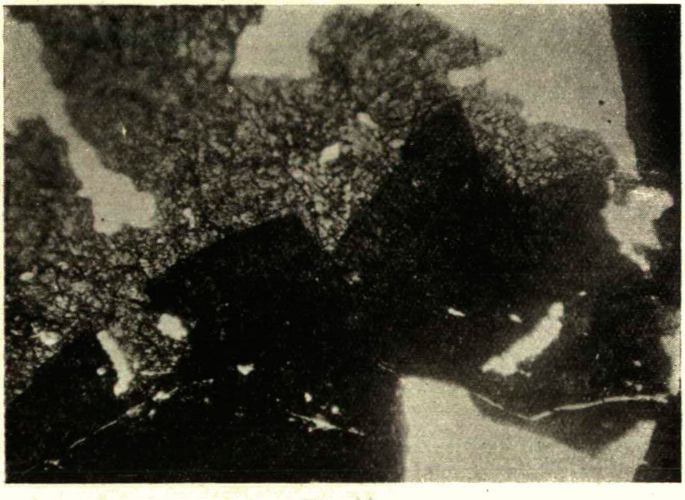

Fig. 12.
Wurtzite rich in iron (dark) with iron-poor sphalerite (gray), × 100

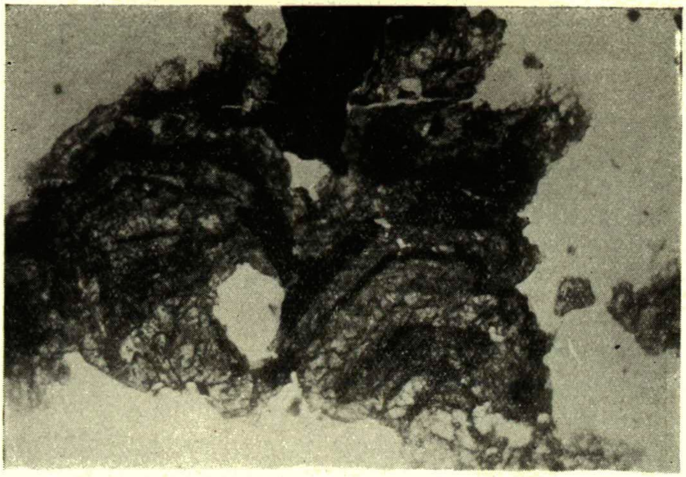

Fig. 13.
Zonal sphalerite, $\times$ 90

From the fragments from which thin sections were prepared polished sections were also made. The latter confirmed all observations made on the thin sections. On the wurtzite crystal groups the chalcopyrite inclusion rows extending along the single crystals forming fanlike not extensive branches, as well as the inclusion rows running parallel to the (0001) cleavage plane in an almost perpendicular direction to the former, can be well detected. The latter as shown on Microphot. 6. succeed each other in particularly dense rows. In the sphalerite crystal groups intergrown parallel to the wurtzite crystal ones the inclusion system formed regularly but in a manner diverging from that of the wurtzite is very conspicious. The shape of the sphalerite tetrahedrons can always easily be distinguished on the basis of the inclusions above wurtzite crystal group (Microphot. 14 and 15). Here it is also immediately plausible that not dismixture of the sphalerite-chalcopyrite system, but an oriented mineral intergrowth of two, with the wurtzite three, minerals is involved. An interesting polished section is that in which the wurtzite and the sphalerite orientally intergrown with it mostly do not contain inclusions, whereas the crystal group is limited by a chalcopyrite inclusion system running parallel with the cleavage plane (Microphot. 16). In this case also the shape of the sphalerite tetrahedron growing orientally, the inclusion row extending on the mutual (0001) (111) plane is very cleanly detectable. The crystal group surrounded by the chalcopyrite continued to develop and after a time another chalcopyrite inclusion system encircled it, a phantom crystal formed.

Besides the sections running parallel, or almost parallel, to the c axis others perpendicular to it were also prepared. The cross-section is a hexagon in which the parts containing more, and those containing less chalcopyrite inclusions alternate zonally (Microphot. 17). This zonal distribution of the inclusions can also be well detected in the polished sections. If the section was prepared obliquely to the *c* axis the hexagon was distorted (Microphot. 18, 19 and 20).

The two crystallised variations of ZnS can only occur beside each other under certain, definite, physico-chemical conditions according to A l l e n and C r e n s h a w, only if the solution contains at a temperature of 330—220°C 4—0.1 per cent of sulphuric acid. When the acid content is higher and the temperature is lower wurtzite, when the former is lower and the latter is higher sphalerite crystals form. It is not probable that our solutions contained any free sulphuric acid, however, it may be assumed that HF was the cause of the acid character of the solution. This

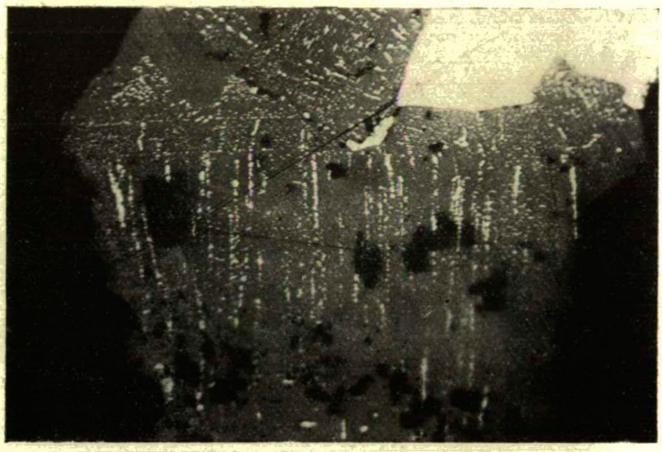

Fig. 14.
Wurtzite-sphalerite in oriented intergrowth, × 400

Fig. 15.
Wurtzite-sphalerite in oriented intergrowth, × 420

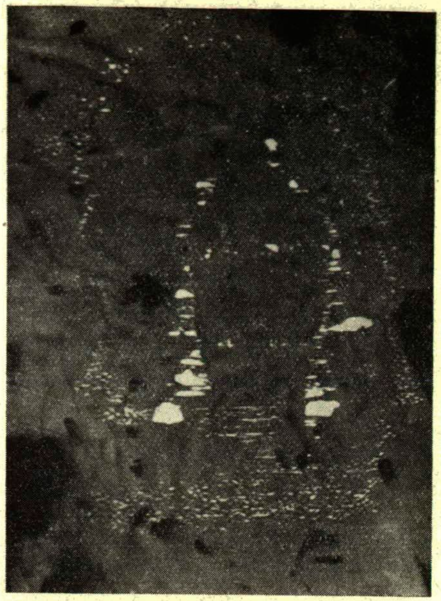

Fig. 16.
Wurtzite-sphalerite with chalcopyrite border, $\times$ 450

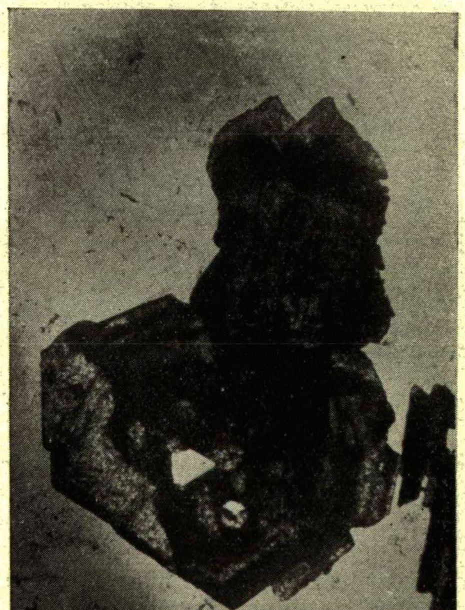

Fig. 17.
Wurtzite crystal groups parallel and perpendicular, respectively to the vertical axis, with oriented chalcopyrite inclusions, $\times$ 110

Fig. 18.
Zonal wurtzite, parallel to the plane 0001, × 450

Fig. 19.
Zonal wurtzite, parallel to the plane 0001, × 340

suggestion is supported by the fact that in the younger crystalline quartz, surrounding the ores, very well developed small fluorite crystals, octahedrons, octahedral crystal groups were found as inclusions (Microphot. 21). These crystal groups were sometimes deposited on wurtzite (Microphot. 22) and sometimes on the older galena replaced by the quartz (Microphot. 23). The row of fluorite crystal inclusions (Microphot. 24) following the lines of the quartz crystal growth which are enclosed by the growing quartz crystals is very fine. Considering that the free acid content of the solution is not known wide limits ranging between 330—220°C must be set for the beginning of the ore-forming probably, however, it occurs in the vicinity of the lower temperature ranges. Consequently the ore formation began in the mesothermal period. When, owing to the formation of fluorite, the free acid content of the solution vanished the separation of wurtzite ceased, among the younger ores forming at lower temperatures this mineral can no more be found.

Wurtzite is not a wide distributed mineral of Gyöngyösoroszi, so far it was only found at some points of the main lode, the Károly lode. It is more or less frequently associated with intergrown fluorite crystal microscopi-

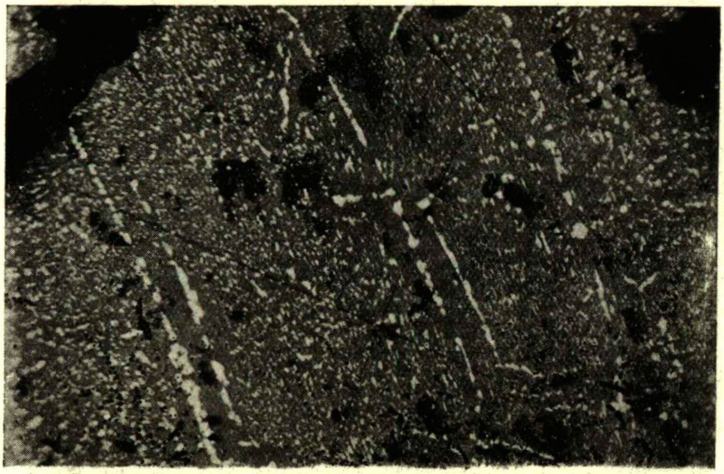

Fig. 20.
Oriented intergrowth of wurtzite and chalcopyrite, × 500

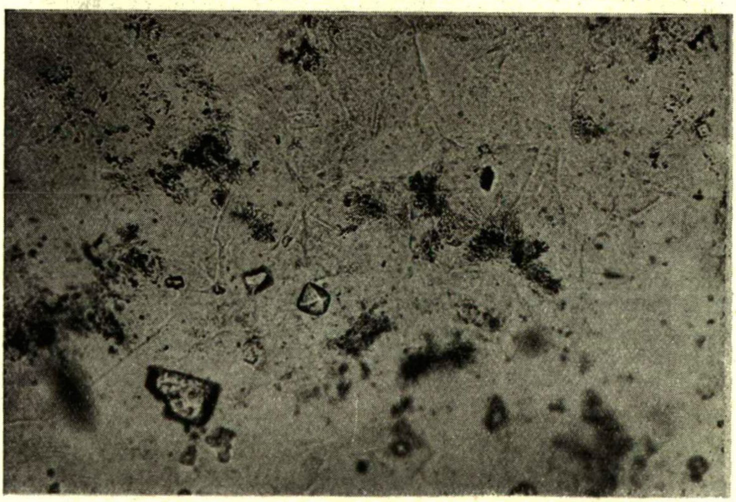

Fig. 21.
Fluorite crystals in quartz, × 170

cally small in size. Moreover fluorite cannot only be detected among the lodes, but also fairly abundantly[8] among the minerals of the cavities of the tufaceous agglomerates and the amygdaloidic andesite minerals of the adit indicating that a considerable amount of the HF contained in the residual solution was already bound before the formation of the ore lodes.

Where the residual solution did not contain fluorine acid the first generation of ZnS separated as black coloured sphalerite containing a

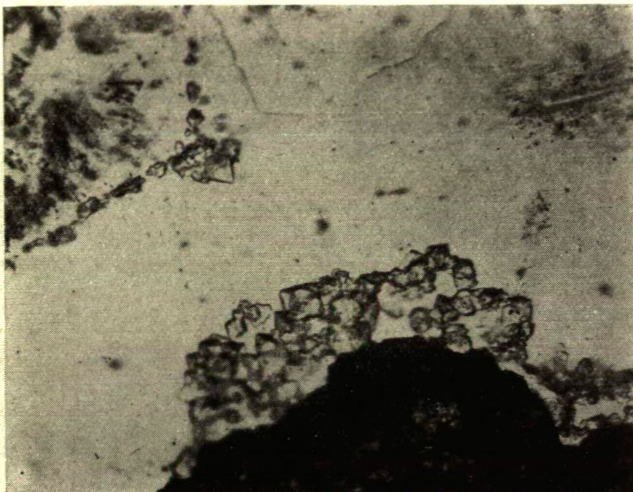

Fig. 22.
Fluorite crystals on wurtzite, $\times$ 170

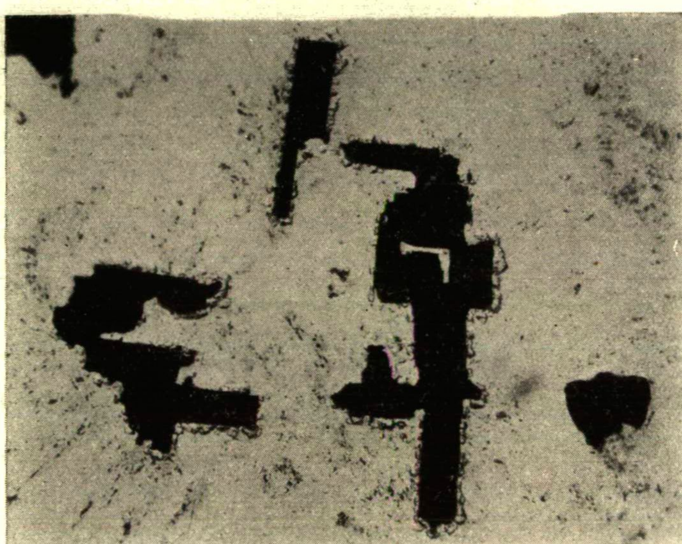

Fig. 23.
Fluorite crystals on galena replaced by quartz, $\times$ 125

large amount of iron and many chalcopyrite inclusions. Under the microscope it shows reddish-brownish internal reflexes. Undoubtedly the inclusions are partly the product of dismixture, however, the two ores can also be found separating rhythmically, the coherent chalcopyrite rows overgrow it orientally and change according to the substance of the sphalerite crystal (Microphot. 25). However, the overwhelming majority of the inclusi-

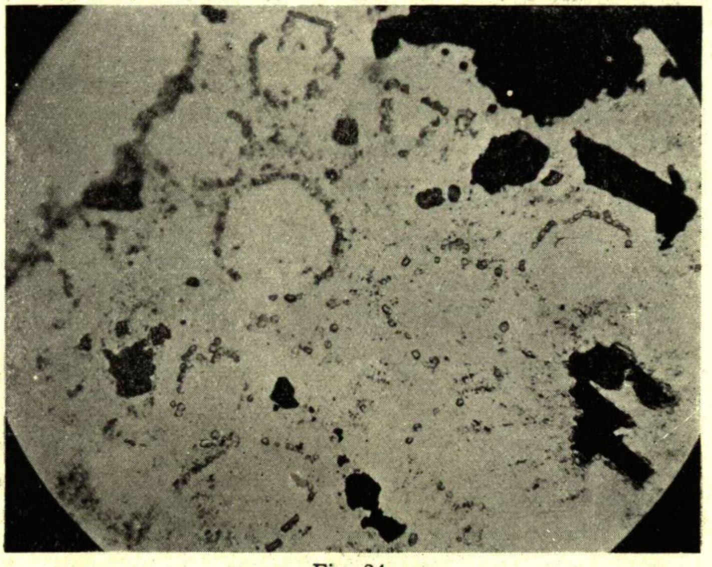

Fig. 24.
Fluorite crystals oriented along the periphery of quartz crystals, $\times$ 80

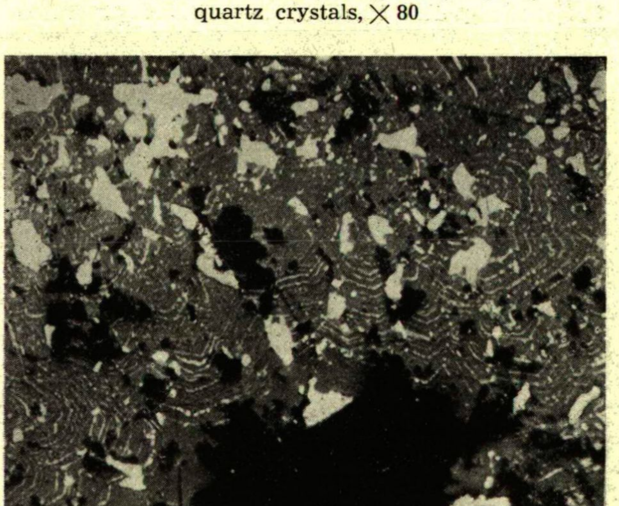

Fig. 25.
Rhythmically oriented separation of sphalerite and chalcopyrite, $\times$ 250

ons are chalcopyrite fragments replaced by the recrystallised sphalerite. The remnants of the original ore can sometimes be found regularly, and sometimes quite irregularly localised in the sphalerite surrounding, and at the same time, replacing it (Microphot. 26, 27). In the former case the differently orientated original chalcopyrite granule boundaries (Microphot. 27) can be reconstructed. Around the larger chalcopyrite islands the replacing sphalerite very frequently contains oriented chalcopyrite

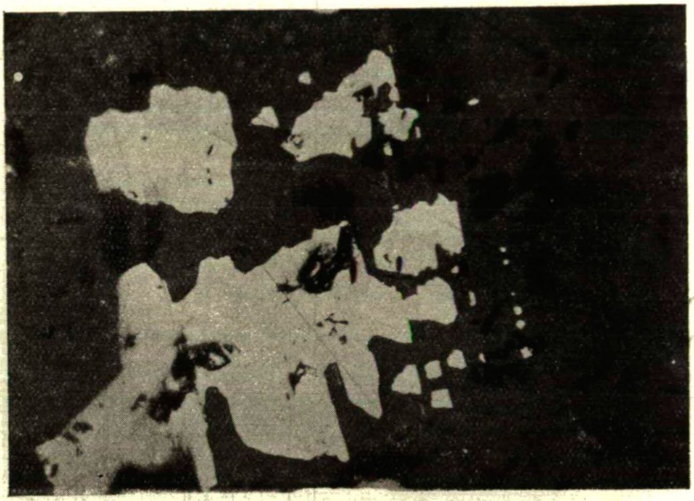

Fig 26.
Sphalerite replacing chalcopyrite, × 340

Fig. 27.
Sphalerite replacing chalcopyrite, × 260

inclusion rows which can only be well detected in oil immersion, these also represent remnants of the replaced chalcopyrite.

Besides the chalcopyrite older than the sphalerite by which it is replaced and that formed simultaneously with the sphalerite which separated rhythmically with it, the older sphalerites which are rich in iron also contain younger chalcopyrite. This chalcopyrite permeates in thin veins taking an irregular course and widening at some places the sphalerite probably following the fissures of the ore resulting from tectonic effects. In these chalcopyrite ores the ore sometimes alternates with stannite. The grey ore possessing a relatively weak reflection capacity did not show pleochroism, but between crossed nicols the interference phenomenon can be detected. By means of the spectroscope the stannum contained in the black sphalerite to which it is bound is well visible (Microphot. 28).

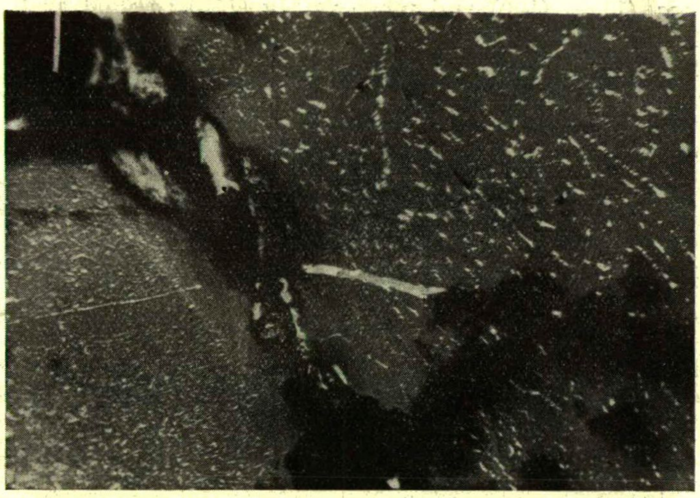

Fig. 28.
Stannite with chalcopyrite, $\times$ 500

Apart from chalcopyrite, both wurtzite and sphalerite, often also contain remnants of the older galena replaced by them. Microphot. 25 illustrates well the galena remnants replaced by the two rhythmically separated ores sphalerite and chalcopyrite. It seems very likely that after pyrite, the strongly corroded remnants of which can be observed in all so far mentioned ores, the first galena generation separated, however, the greater part of this galena was replaced by chalcopyrite and sphalerite (wurtzite). These sulphide ores formed at higher temperatures only in small amounts were replaced by quartz which separated in great masses. The replacement followed the cleavage planes of the sulphides.

Hence, at higher temperatures only small amounts of ore separate therefore in many of the hitherto exposed ore lode sections, they cannot be found only later, after the temperature of the solution decreased ores formed in larger quantities.

Sphalerite which separated as second generation is the dominating ore of the lodes, it has a brownish-yellow colour and its iron content is appreciably lower than that of the former. This sphalerite composes with galena and with very little or no chalcopyrite, either coarsely granular groups in thinner or thicker cords in quartzous gangue or the sphalerite is ingrown as scattered rounded rhombohedral shaped grains in quartz and calcite respectively. Among these intergrown crystals those showing a zonal structure are frequent, the younger layers can be separated from the somewhat darker coloured nucleus. This sphalerite, which is younger and contains less iron than that described above exhibits under the microscope a light, yellowish-brownish, internal reflex having far less chalcopyrite inclusions, these occurring in it are chalcopyrite remnants replaced by sphalerite.

Its little crystals, less than one cm in size, can often be found in smaller cavities overgrown on quartz, planes (111) dominate on them, besides them planes $(1\bar{1}1)$, (100) and more rarely (110) as well as (311) can be detected the latter shape is incompletely developed and has only a few planes. The greater part of the crystals are twins. The angular rubble sphalerite contained in the quartz of the brecciated ore frequently occurring in the Károly lode fell into fragments at the crystallisation of the large amounts of silicic acid which had separated in the gel state.

The chemical analyses of wurtzite (I), the black, older sphalerite (II) and the quantitatively dominating lighter young sphalerite (III) yielded the following results:

	I. a.	I. b.	II.	III. a.	III. b.
Zn	29,71 %	42,99 %	55,87 %	46,54 %	59,78 %
Cd	0,11	not determ.	0,48	0,29	not determ.
Fe	3,12	6,52	3,85	1,92	2,58
Mn	0,15	trace	0,10	0,10	0,69
Cu	0,62	0,78	0,31	0,07	0,10
Pb	9,73	10,77	3,95	4,03	1,66
As	—	—	—	trace	trace
S	18,30	26,96	31,21	24,72	32,87
SiO_2	37,75	12,54	3,89	21,84	1,72
	99,47 %	100,56 %	99,66 %	99,51 %	99,40 %

Subtracting the insoluble SiO_2 and calculating the sulphide residual for 100 per cent the following results are obtained:

	I. a.	I. b.	II.	III. a.	III. b.
Zn	48,14 %	48,86 %	58,35 %	59,94 %	61,21 %
Cd	0,17	—	0,50	0,37	—
Fe	5,05	7,40	4,02	2,47	2,64
Mn	0,24	—	0,10	0,12	0,70
Cu	1,00	0,88	0,32	0,09	0,10
Pb	15,75	12,23	4,12	5,18	1,69
S	29,65	30,63	32,59	31,83	33,66
	100,00 %	100,00 %	100,00 %	100,00 %	100,00 %

Besides these trace elements which could also be identified by means of chemical analyses the spectroscopical examination still showed the following:

Ag in all three samples, in the following intensity sequel: II., III., I.

Sn in all three samples, in the following intensity sequel: II., far weaker III, I.

Considering that also under the microscope stannine could only be detected in the older sphalerite, marked with II., the results of the spectroscopical investigation are in this case too in good agreement with our observations.

The younger, light, sphalerite fluoresces with a nice dark violet colour, on the action of ultraviolet rays. In the case of dark sphalerite and wurtzite, owing to their high iron content, the fluorescence phenomenon is very slight or fails to occur.

The first generation of the other dominating ore of the lodes, galena, is as already mentioned partly older than wurtzite and black sphalerite. Its amount is also in this case insignificant, the two variation of ZnS and quartz replace it. Both wurtzite and the older sphalerite very frequently contain larger or smaller galena remnants showing already partly advanced replacement, this explains why the two zinc ores always contain lead in considerable amounts. (Microphot. 29). On chalcographical etching this older galena showed a nice zonal structure. In two sections of these galena remnants replaced by the older sphalerite very tiny gold grains, several microns in size, could be detected. (Microphot. 30). Consequently, the separation of gold began simultaneously with the formation of the sulphide ore. The older galena also already contains bournonite in small patches it is, however, far rarer in this ore than in the dominating younger galena.

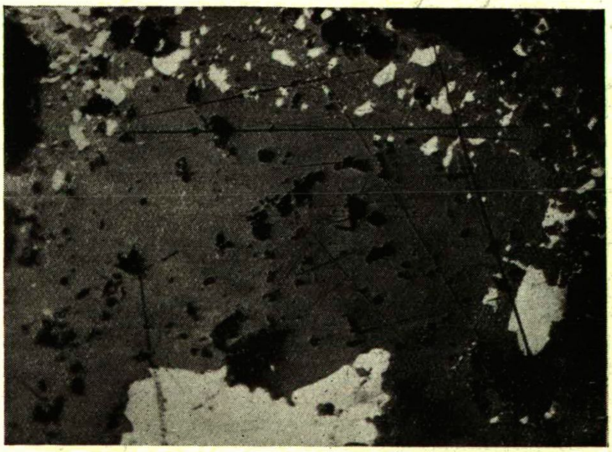

Fig. 29.

Sphalerite replacing galena with chalcopyrite inclusion rows of the same age than the latter, $\times$ 170

The latter associated with light sphalerite is one of the main ores of the mining district, it occurs in the gangue in dispersed granules or as independent thinner veins composing smaller nests, in the latter case it forms coarsely granular, or radial-crystalline masses. In the polished sections prepared from this galena parallel dot rows due to the strikingly fine mother liquor inclusion mentioned by Sztrókay are frequent. The separation of galena began before that of the younger sphalerite — this

Fig. 30.
Gold in galena replaced by sphalerite, × 300

fact has also been established by Sztrókay — the sphalerite contains remnants of the galena displaced by it, hence it also contains lead in considerable amounts, whereas the large zinc content of galena (s. analysis) is due to the sphalerite replacing it developing along the mother liquor inclusions mentioned above.

In the smaller cavities very fine overgrown galena crystals can be found. The crystals are usually combinations of planes (111) and (100) the former is the dominating one, the latter blunts the peaks of the former, less frequently small hexahedrons also occur. A small galena crystal ingrown in granular calcite is apparently a hexagon tabular is a deficiently developed contact twin which flattened according to one of the axis of the trigonal symmetry of the two cube-octahedrons developed in equilibrium.

The galena is replaced by sphalerite, chalcopyrite, and by the bournonite formed at expense of the substance of the galena, as well as by the jamesonite and semseyite. The granules of bournonite are always arranged in very fine twin lamellae they occur fairly frequently. This mineral was also found in small overgrown twin crystals. The crystals are about one mm in size and are overgrown on quartz. Jamesonite needle aggregates appear at the borders of the galena, sometimes the jamesonite formed at the expense of the galena completely surrounds some single galena granule. A fanlike crystal group overgrown at the edge of a small galena crystal in the brecciated ore also occurred, on the basis of its reflection and interference we identified it as semseyite. In the younger galena gold could not be found, neither could the boulangerite mentioned by Sztrókay, nor the miargyrite marked by him with note of interrogation be identified. In all sections traces of a commencing decomposition of the galena could be observed it is permeated by thin cerussite veins containing one or more covellite plates. The result of the analyses of the younger galena are as follows:

	I.	II.
Pb	71.54 %	75,42 %
Zn	0,64	7,69
Fe	0,91	0,12
Cu	trace	0,75
Mn	trace	—
Sb	0,20	0,48
S	16,92	15,51
SiO_2	0,45	0,25
	99,66 %	100,22 %

The material for the analyses was taken from a coarsely radial pure small galena lode. The Sb content is due to the not negligable lead-antimony sulphides.

The third, less frequent, ore of the mining district is chalcopyrite also occurring in two generations an older and a younger one. It always can only be detected as small granules or patches, it has not yet been found as a coherent lode filling, like sphalerite or galena. It is partly older than sphalerite and partly it separated at the same time as the latter. It frequently contains badly corroded granules of the older pyrite. The polished sections of chalcopyrite always exhibited between crossed nicols a twin lamellae structure even the electrographically etched granules showed a very thin polysynthetic twin lamellae structure. Intergrown in the quartz of the lode scattered, well developed, sphenodic small crystals also occur, whilst more recently larger overgrown curved sphenoidic crystals with blurred planes could be observed in the southern lodes of Hidegkut 2 and also in those of Kőkut. At the edge of the chalcopyrite granules in some of the polished sections a thin chalcocite band containing small covellite lamellae occurred.

Among the sulphide ores crystals of pyrite were the first to separate. Their intergrown hexahedral crystals can already frequently be seen in the accessory rocks and also at the edges of the lodes which are in contact with the latter. In the first generation of all sulphide ores the strongly resorbed remnants of these crystals can always be formed. The second pyrite generatio consists of small compact granules it does not occur in the Károly lode in larger masses or as independent nests, as thin cords in dispersed patches, however, it is not infrequent. It encrusts sphalerite, quartz and calcite and can be found overgrown on them as small hexahedral, pentagonal dodecahedral crystals or crystal groups. In the calcite filling of the Károly lode compact, quite gellike pyrite, forms veins attaining one cm in size which often limit the light coloured calcite from the brown- dark brownish one stained by MnO_2 [2]. The interior of the pyrite exhibiting a gel structure, patchings shining in bright interference colours often can be found. Sztrókay assumes that these more compact small granules which are easier to polish than the gellike pyrite are, owing to their As or Sb content, pyrite granules exhibiting an anomalic optical character, in the view of the author they are marcasite crystals. The more so as marcasite occurs extensively in our mining district. Its crystal aggregates with their fine twin crystals can often be detected in our sections. It represents an interesting phenomenon when its thin needle crystals are overgrown on idiomorphous hexahedral pyrite crystals, quasi forming a frame around them.

On overgrown sphalerite crystals arsenopyrite crystals hardly one mm in size surrounded by planes (110), (001) and (101) occurred. Our sections did not show this ore.

Not taking quartz and its varieties, which can now be found in dominating amounts in this mining district, furthermore calcite, also occurring in large amounts, into consideration only laumontite and cerussite observed as secondary minerals will still be mentioned.

Laumontite was detected on overgrown small quartz crystals clear as water at the 600th m of the adit. On the small crystals 2—4 mm in size we succeeded in establishing besides the dominating (110) planes still those of (111) and (001).

Small cerussite crystals were found in the hollows of a sphalerite pyrite samples. On the crystals as clear as water the following forms could be observed: (001), (010), (110) and (021). They are slightly alongated hexagonal tabular shaped crystals with the dominating base.

According to our grouping the following minerals could so far be observed at Gyöngyösoroszi:

Ores: pyrite I, galena I, gold, chalcopyrite I, sphalerite I, wurtzite, stannite, bournonite, *galena II, sphalerite II,* chalcopyrite II, marcasite, pyrite II, jamesonite, semseyite, arsenopyrite, tetrahedrite, antimonite.

(We did not succeeded in finding the boulangerite and miargyrite mentioned by Sztrókay.)

Non ore minerals: *quartzvarieties* (quartzite, chalcedony, jasper, rock crystal, amethyst), opal, *calcite,* dolomite, fluorite, pennine, barite, coelestite, gypsum I, laumontite. Among the minerals of the tufaceous agglomerate: adularia.

The minerals of the oxidized zone: chalcocite, covellite, cerussite, sulphur, gypsum II.

The chemical elements taking part in the formation of the minerals of the ore lodes: *O Si Ca S C Fe Zn Pb* Mn Mg Cu H F Cd Ba Al Sr P K As Ag Au.

Oocurring in traces which can only be detected spectroscopically Sn Mo.

SUMMARY:

1. The substance of the residual solution containing large amounts of silicic acid separated in the older longitudional ore lodes rhythmically, but coherently. The smaller part of the separated material is of mezo-, the greater part of epithermal origin. The silicic acid separating as gel and crystallising slowly partly broke up the earlier crystallised ore embedding its fragments and partly displacing it. 2. The longitudional ore lodes belong to the row of the zinc lead ore lodes (Selmecbánya, Felsőbánya, Kapnikbánya etc.) which contain gold and silver and are wide distributed among the subvulcanic ore formations bound to the young tertiary mountain ranges, unfortunately, however, they are far less rich and do not resemble in anyway the ore occurrence of Nagybörzsöny.

The extremely variable crystals of calcite and crystallised quartz in which solid and liquid inclusions occur so abundantly seem worthy of further investigation.

Analyses and microphotographs by Gy. Grasselly.

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